

# Thickness Dependence of the Reorientation Phase Transition

David Clarke

*Johns Hopkins University, Department of Physics and Astronomy,  
3400 N. Charles St., Baltimore, Maryland 21218*

This report examines the thickness dependence of the reorientation phase transition in ferromagnetic films with perpendicular anisotropy. That is, we find the exact boundary of metastability of uniformly magnetized in-plane states as the solution to a set of transcendental algebraic equations, and find the profile of the initial instability in the magnetization in the direction normal to the plane of the film. In general, this instability occurs at a finite wave number  $k$ . We determine the dependence of  $k$  on the film thickness.

## I. INTRODUCTION

Experimental and theoretical studies of thin ferromagnetic films with an anisotropy axis perpendicular to the plane of the film have shown a diverse set of magnetic phases. In addition to uniformly magnetized states, strongly modulated (striped) patterns have been observed<sup>1,2,3,4</sup> as the dipolar stray field overcomes the exchange interaction and forces an oscillation of the magnetization. The anisotropy value at which the modulation first occurs for is known as the reorientation phase transition (RPT) point.<sup>5,6,7</sup>

This spontaneous modulation was first modeled by Garel and Doniach,<sup>8</sup> using the assumption that the local magnetization lies fully out of the plane of the film. The magnetization alternates between regions of uniform up and down polarization with width determined by the competition between the ferromagnetic exchange and the dipolar interactions. Refinements of this model have shown that near the RPT the domain structure is a cosine-like modulation of a nearly in-plane state,<sup>6,9</sup> rather than the sharply defined striped domains that occur at higher values of the out-of-plane anisotropy.

Recently,<sup>10</sup> we have found a universal phase diagram for zero-temperature transitions in thin film ferromagnets with perpendicular anisotropy and an external field applied perpendicular to the plane of the sample. In particular, we found analytic solutions for the boundary of metastability of uniformly magnetized phases in thin films. The uniformly up and down states decay along the lines  $h = \mp \kappa$  respectively, while the canted phase decays at the boundary  $(h/\kappa)^2 = 1 + \kappa/\kappa_0$ . These results, however, depend on the smallness of the parameter  $\kappa_0 = t^2/(4\lambda)^2$ , where  $\lambda = \sqrt{2A/\mu_0 M^2}$  is the exchange length. This approximation is not valid for all thin films being studied in current experiments. In particular, it fails in experiments with thermally evaporated thin nickel films,<sup>11</sup> where  $\lambda \approx 20nm$  and  $t$  ranges from  $30nm$  to  $200nm$ .

Three important effects come into play as the film thickness increases. First, the bound charges created on the top and bottom surfaces of the film by out-of-plane magnetization grow further apart, altering the strength of the dipolar interaction. Second, the magnetization may vary across the thickness of the film. Third, the

direction of the in-plane magnetization may vary. The combination of the latter two effects allows the creation of closure domains that decrease the dipolar energy by following the local stray field. We find that at large thicknesses, the lowest energy modes are actually dominated by such domains.

Variational studies of the dependence of the domain structure on the film thickness have been made,<sup>12,13</sup> but have not allowed for variation of the magnetization along the direction normal to the film. While these studies show the correct dependence of the RPT anisotropy on thickness, they predict domain sizes that decrease monotonically with the thickness, opposite the actual trend at large thicknesses.<sup>14</sup>

This report examines the decay of uniform states in a thicker film via a spin wave analysis. We reduce the problem of finding the decay boundary of an in-plane state to a set of transcendental algebraic equations, and find the form of the initial decay modes. Section II reviews the model for the magnetic free energy in a ferromagnetic film. Section III begins the spin wave analysis of the system around a uniformly polarized state, making general arguments about the shape and direction of the lowest energy fluctuations. In Section IV, we find the governing eigenvalue equation for the spin wave modes in the  $z$  direction (normal to the film) and find the general form of solutions to this equation. Section V completes the analysis by finding the mode with minimum energy and determines a set of equations that describe the reorientation phase transition point as a function of the thickness  $t$ .

## II. MODEL FREE ENERGY

The free energy for a thin ferromagnetic film of thickness  $t$  can be separated into local and long-range parts. The local part includes the exchange, uniaxial anisotropy, and Zeeman terms:

$$E_{\text{local}} = \int_{z=-t/2}^{t/2} d^3r (A|\nabla \hat{\mathbf{m}}|^2 - K m_z^2 - \mu_0 M \mathbf{H} \cdot \hat{\mathbf{m}}), \quad (1)$$

where  $\hat{\mathbf{m}} = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$  is the 3-dimensional unit vector pointing along the magnetization. For simplicity, we shall consider the case  $\mathbf{H} = 0$ .

Experimentally, changing the temperature can serve to vary the anisotropy. It has been shown that the effective out-of-plane anisotropy decreases monotonically with increased temperature.<sup>5,6,15</sup>

The long-range part of the energy is due to dipolar interactions:

$$E_{\text{dip}} = \frac{\mu_0 M^2}{8\pi} \int d^3 r \int d^3 r' \frac{\rho(r)\rho(r')}{|\mathbf{r} - \mathbf{r}'|}, \quad (2)$$

where  $\rho(r) = -\nabla \cdot (P(z)\mathbf{m}(\mathbf{r}))$ . The integration runs over all space. Here  $P(z)$  is the profile of the film in the  $z$  direction.  $P(z) = 1$  if  $-t/2 \leq z \leq t/2$  and 0 otherwise.

### III. SPIN WAVE EXPANSION AROUND AN IN-PLANE STATE

Consider the free energy of small fluctuations around an in-plane state in which the magnetization points along the  $y$  axis:  $\theta = \pi/2 + \nu(\mathbf{r})$ ,  $\phi = \pi/2 + \alpha(\mathbf{r})$ , with  $\nu, \alpha$  small. By transforming to Fourier space in the  $xy$ -plane, we can write the energy as an integral over modes that do not interact at second order in the fluctuations:

$$\Delta E = \int \frac{d^2 k}{(2\pi)^2} (\Delta E_1[\nu_{\mathbf{k}}] + \Delta E_2[\alpha_{\mathbf{k}}] + \Delta E_{\text{dip}}[\nu_{\mathbf{k}}, \alpha_{\mathbf{k}}]). \quad (3)$$

This separation of modes is important because it means that the instability at the RPT will occur for a cosine-like fluctuation with a single wavelength. Here

$$\begin{aligned} \frac{\Delta E_{1,\mathbf{k}}}{\mu_0 M^2} &= \int_{-t/2}^{t/2} dz \left[ \frac{\lambda^2}{2} |\nu'_{\mathbf{k}}|^2 + |\nu_{\mathbf{k}}|^2 \left( \frac{\lambda^2 k^2}{2} - \frac{\kappa}{2} \right) \right], \\ \frac{\Delta E_{2,\mathbf{k}}}{\mu_0 M^2} &= \int_{-t/2}^{t/2} dz \left[ \frac{\lambda^2}{2} |\alpha'_{\mathbf{k}}|^2 + |\alpha_{\mathbf{k}}|^2 \frac{\lambda^2 k^2}{2} \right], \end{aligned} \quad (4)$$

and

$$\frac{\Delta E_{\text{dip},\mathbf{k}}}{\mu_0 M^2} = \frac{|k|}{4} \int_{-t/2}^{t/2} dz \int_{-t/2}^{t/2} dz' e^{-|k||z-z'|} f_{\mathbf{k}}(z, z') f_{-\mathbf{k}}(z', z), \quad (5)$$

where

$$f_{\mathbf{k}}(z, z') = \text{sgn}(z - z') \nu_{\mathbf{k}}(z) - i \frac{k_x}{|k|} \alpha_{\mathbf{k}}(z). \quad (6)$$

Primes on  $\nu$  and  $\alpha$  denote  $z$ -derivatives. We define the effective anisotropy  $\kappa = (K - \mu_0 M^2)/(\mu_0 M^2/2)$  as the natural anisotropy less the uniform part of the dipolar interaction.

Note that the dipolar energy couples symmetric fluctuations in  $\nu$  to antisymmetric  $\alpha$  fluctuations and vice-versa. In general, fluctuations in the azimuthal angle  $\phi$  allow a gain in energy by compensating for the magnetic charges created by fluctuations in the polar angle  $\theta$ .

It is these azimuthal fluctuations that cause the lowest energy mode to have its wave vector perpendicular to the initial magnetization (i.e. in the  $x$ -direction).

To see this, consider a mode such that the wave vector of the oscillation is at some angle  $\beta$  to the  $x$ -axis, with a profile  $\{\alpha_{\mathbf{k}}^{(0)}, \nu_{\mathbf{k}}^{(0)}\}$  for the azimuthal and polar fluctuations. For any such mode, we can find a corresponding fluctuation that has exactly the same dipolar energy but has its wave vector along the  $x$ -axis. Namely,  $\{\alpha_{\mathbf{k}}^{(1)}, \nu_{\mathbf{k}}^{(1)}\} = \{\alpha_{\mathbf{k}}^{(0)} \cos \beta, \nu_{\mathbf{k}}^{(0)}\}$ .

Further, since the azimuthal part of this new fluctuation is smaller by a factor of  $\cos \beta$ , it has a lower cost in the exchange energy. As a result, the difference in energy between the two types of fluctuations,

$$E_{\mathbf{k}}^{(0)} - E_{\mathbf{k}}^{(1)} = \sin^2 \beta_0 \int_{-t/2}^{t/2} dz \left[ \frac{\lambda^2}{2} |\alpha'_{\mathbf{k}}^{(0)}|^2 + |\alpha_{\mathbf{k}}^{(0)}|^2 \frac{\lambda^2 k^2}{2} \right] \quad (7)$$

is a nonnegative quantity.

Since this is true of any profile for the azimuthal and polar fluctuations, it must be that the mode with the lowest energy cost has its wave vector oriented along the  $x$ -axis, perpendicular to the original direction of the magnetization.

The exception to the above is when the wave vector of the oscillation lies along the  $y$ -axis, since the azimuthal fluctuation is then zero. In this case, however, no compensation can be made for the charges created by the fluctuations in  $\theta$ , so the energy will be inherently higher.

### IV. VERTICAL SPIN WAVE EIGENMODES

Given that the lowest energy mode occurs with a wave vector along the  $x$ -axis, we work only in the subspace  $k_y = 0$ . The energy in this subspace is diagonalized by modes that obey the eigenvalue (Lagrange) equations:

$$\begin{aligned} \gamma \alpha_{\mathbf{k}} &= -\lambda^2 \alpha_{\mathbf{k}}'' + k^2 \lambda^2 \alpha_{\mathbf{k}} \\ &+ \frac{k}{2} \int_{-t/2}^{t/2} dz' e^{-|k||z-z'|} f_{-\mathbf{k}}(z', z) \end{aligned}$$

and

$$\begin{aligned} \gamma \nu_{\mathbf{k}} &= -\lambda^2 \nu_{\mathbf{k}}'' + (k^2 \lambda^2 - \kappa) \nu_{\mathbf{k}} \\ &+ \frac{|k|}{2} \int_{-t/2}^{t/2} dz' e^{-|k||z-z'|} i \text{sgn}(z' - z) f_{-\mathbf{k}}(z', z) \end{aligned} \quad (8)$$

with  $\alpha'(-t/2) = \alpha'(t/2) = \nu'(-t/2) = \nu'(t/2) = 0$  induced by the boundary terms used in deriving the above equations. The energy of each mode is proportional to its eigenvalue  $\gamma$ . For a given value of  $t$ , the reorientation phase transition will occur at the  $\kappa$  value when the lowest eigenvalue crosses zero, and some mode becomes soft. That is, at  $\kappa$  such that  $\gamma = d\gamma/dk = 0$  for some mode.

Note that the integral kernel in Eq. (8) is the solution to the differential equation  $G''(z) - k^2 G(z) = \delta(z)$ , which has constant coefficients. Since (8) is linear, and all the

other terms in (8) have constant coefficients as well, we would expect Eq. (8) to be solved by

$$\alpha_{\mathbf{k}} = \sum \alpha_{i,\mathbf{k}} e^{q_i z}, \quad \nu_{\mathbf{k}} = \sum \nu_{i,\mathbf{k}} e^{q_i z}, \quad (9)$$

where  $\{\nu_{i,\mathbf{k}}\}$ ,  $\{\alpha_{i,\mathbf{k}}\}$ , and  $q_i(k)$  are constants.

This ansatz works so long as the  $\{q_i\}$  are the solutions to the equation

$$a(a - \gamma)(a - \gamma - \kappa) = (\kappa + 1)k^2\lambda^2, \quad (10)$$

where  $a = (-q^2 + k^2)\lambda^2$ , and the conditions

$$\begin{aligned} 0 &= -iq_i k \nu_{i,\mathbf{k}} - (a_i^2 - \gamma a_i + k^2)\alpha_{i,\mathbf{k}}, \\ 0 &= \sum \frac{e^{q_i t/2}}{|k| - q_i} (i|k|\nu_{i,\mathbf{k}} + k\alpha_{i,\mathbf{k}}), \\ 0 &= \sum \frac{e^{-q_i t/2}}{|k| + q_i} (i|k|\nu_{i,\mathbf{k}} - k\alpha_{i,\mathbf{k}}). \end{aligned} \quad (11)$$

are satisfied. Note that there are six (not necessarily distinct) values for  $q$ , separated into three pairs  $q = \pm\sqrt{k^2 - a/\lambda^2}$ .

The boundary conditions on the top and bottom surfaces lead to

$$\begin{aligned} 0 &= \sum q_i e^{-q_i t/2} \alpha_{i,\mathbf{k}} = \sum q_i e^{q_i t/2} \alpha_{i,\mathbf{k}}, \\ 0 &= \sum q_i e^{-q_i t/2} \nu_{i,\mathbf{k}} = \sum q_i e^{q_i t/2} \nu_{i,\mathbf{k}}. \end{aligned} \quad (12)$$

These boundary conditions, combined with Eqs. (8) and (10) are symmetric in the  $z$ -direction, allowing the separation of  $\nu$  and  $\alpha$  into modes with definite parity in the  $z$ -direction. A symmetric  $\nu$  couples only to an antisymmetric  $\alpha$  and vice-versa.

## V. LOWEST ENERGY EIGENMODE AND THE RPT

In the thinnest films,  $\nu$  is uniform across the thickness, and  $\alpha$  is zero. As the thickness increases, we expect the parity of the lowest energy eigenmode to remain the same. The reason that a symmetric  $\nu$  is favored is that the charges created on the top and bottom surfaces will be of opposite sign. This configuration has a lower energy cost than one with the same charges on the top and bottom surfaces, as would be the case for an antisymmetric  $\nu$ .

If  $\nu$  remains symmetric, then

$$\alpha_{\mathbf{k}} = \sum \tilde{\alpha}_{i,\mathbf{k}} \sinh(q_i z), \quad \nu_{\mathbf{k}} = \sum \tilde{\nu}_{i,\mathbf{k}} \cosh(q_i z), \quad (13)$$

where the sum is over the three possible  $a_i$ , the positive branch is taken for  $q_i$ , and  $\tilde{\alpha}_{i,\mathbf{k}}$  and  $\tilde{\nu}_{i,\mathbf{k}}$  are the appropriately symmetrized combinations of  $\alpha_{i,\mathbf{k}}$  and  $\nu_{i,\mathbf{k}}$ .

In this case, the constraint equations (11) and (12) reduce to the set

$$0 = \sum \left( \frac{|k|}{q_i} (a_i - \gamma + 1) \cosh \frac{q_i t}{2} + (a_i - \gamma) \sinh \frac{q_i t}{2} \right) \tilde{\alpha}_{i,\mathbf{k}},$$

$$\begin{aligned} 0 &= \sum \left( q_i \cosh \frac{q_i t}{2} \right) \tilde{\alpha}_{i,\mathbf{k}}, \\ 0 &= \sum \left( (a_i^2 - \gamma a_i + k^2 \lambda^2) \sinh \frac{q_i t}{2} \right) \tilde{\alpha}_{i,\mathbf{k}}. \end{aligned} \quad (14)$$

In order to find the reorientation phase transition bound-

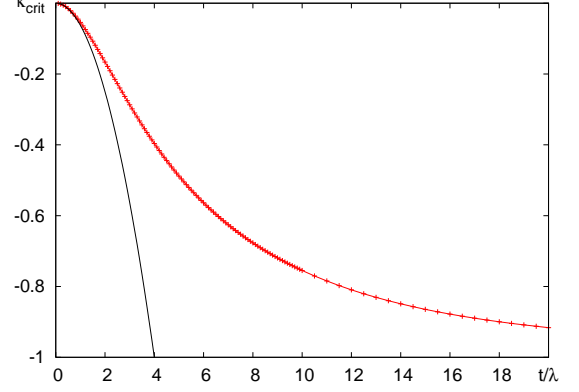


FIG. 1: Dependence of the reorientation phase transition anisotropy  $\kappa_{\text{crit}}$  on the film thickness. The line with + marks is the exact solution, while the solid line is the thin film limit  $\kappa_{\text{crit}} = -(t/4\lambda)^2$

ary, we need not solve for  $\tilde{\alpha}_{i,\mathbf{k}}$ , but we may rather note that in order for the above system to have a non-trivial solution, the determinant of the constraint conditions must be zero. Combining this with the condition  $\gamma = d\gamma/dk = 0$  for the RPT, we find that

$$\begin{aligned} 0 &= \epsilon_{lmn} D_{lmn}, \\ 0 &= \epsilon_{lmn} \frac{d}{dk} D_{lmn}, \end{aligned} \quad (15)$$

where

$$\begin{aligned} D_{lmn} &= \left( \frac{|k|}{q_l} (a_l + 1) \cosh \frac{q_l t}{2} + a_l \sinh \frac{q_l t}{2} \right) \\ &\times \left( q_m \cosh \frac{q_m t}{2} \right) \left( (a_n^2 + k^2 \lambda^2) \sinh \frac{q_n t}{2} \right) \end{aligned} \quad (16)$$

Together with Eq. (10) for the  $q$  values, this completes the system of equations necessary to find both the  $\kappa_{\text{crit}}$  value of the RPT and the initial wavenumber  $k$  of the initial oscillation. Since these equations are transcendental, no further analytic progress may be made. A plot of the numerically determined solution to these equations is found in Figs. 1 and 2, along with a comparison to the previously determined thin-film limit<sup>10</sup>. The thin film limit  $\kappa_{\text{crit}} \rightarrow -(t/4\lambda)^2$ ,  $k \rightarrow t/4\lambda^2$  can be seen to work well for thicknesses  $t \lesssim 2\lambda$ . As the thickness increases, the initial wavenumber reaches a maximum (when  $t/\lambda = 3.7 \pm .1$ ), and decreases to zero (uniform rotation) for infinite thickness. For large thicknesses, the wavenumber goes like  $\pi/t$ , which is consistent with the linear dependence of domain size on thickness in thicker films.<sup>14</sup> The critical value of the effective

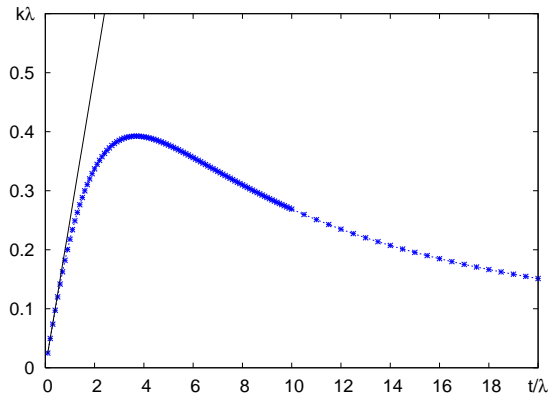


FIG. 2: Dependence of the instability wavenumber  $k$  on thickness. The starred line is the exact solution. The solid line is the thin film limit  $k \rightarrow t/4\lambda^2$ .

anisotropy changes monotonically from its thin film limit to the bulk limit  $\kappa = -1$  at  $t = \infty$ .

The dependence of the entire canted phase boundary on thickness may be derived in a similar fashion. Further complications arise, however, as the characteristic equation (10) loses the symmetry that allows us to solve it as a cubic, and becomes fully sixth order. Further, when the initial magnetization is tilted, the mode with the lowest eigenvalue will no longer have its wavevector parallel to the  $x$ -axis.

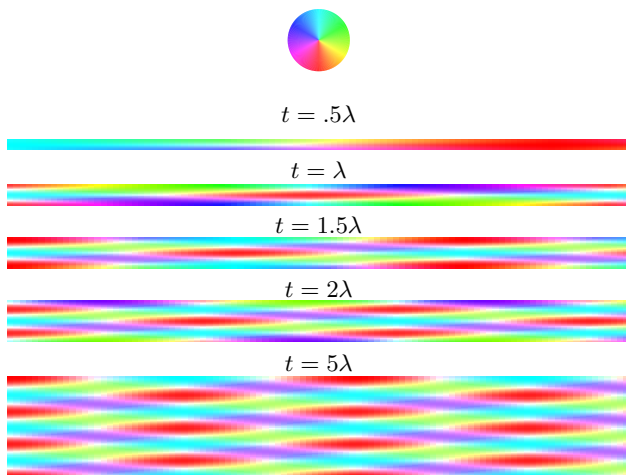


FIG. 3: First unstable modes for various thicknesses. The film is shown in profile, such that magnetization is mostly into the page, with small fluctuations in the directions indicated by the color wheel. The modes are periodic in the  $x$ -direction, which runs across the page. A section  $30\lambda$  wide is shown.

## VI. CONCLUSIONS

In this report, we have developed the thickness dependence of the reorientation phase transition, deriving values for  $\kappa_{\text{crit}}(t)$  and  $k(t)$  that may be directly compared with experiment. In so doing, we have connected recently derived thin film limits with the well-known linear dependence of domain size on thickness in thicker films.

Fig. 3 shows the fluctuation patterns in the modes that first become unstable for a variety of thicknesses. In the thinnest films, the assumption that the magnetization is uniform across the thickness of the film is valid. The azimuthal angle is nearly zero everywhere. As the thickness increases, however, this assumption breaks down, leading to a drastically different landscape of magnetization as closure domains form near the upper and lower surfaces of the film. For very thick films, the lowest energy mode can have many repetitions of this closure behavior.

As the anisotropy increases well beyond the RPT point, it is likely that many of these repetitions would be forced out, to accommodate the demand for more vertical magnetization. However, at the RPT point itself, this leads for very thick films to an essentially periodic structure in the  $z$ -direction.

Since the transition away from the in-plane state as the anisotropy increases is continuous,<sup>6,10</sup> the ground state of a magnetic film that is just beyond the transition point should look much like the modes in Fig. 3 that first become unstable. For such films we predict a strong, nearly oscillatory thickness dependence of the stray magnetic field, as the surface of the film becomes dominated alternately by out-of-plane magnetization, as for  $t = 1.5\lambda$  and  $t = 5\lambda$ , and in-plane closure domains, as for  $t = \lambda$  and  $t = 2\lambda$ . This effect may be observable with magnetic force microscopy.

## VII. ACKNOWLEDGEMENTS

Thanks to M. McEvoy, O. A. Tretiakov and O. Tchernyshyov for helpful discussions, and to S.H. Lee, F. Q. Zhu, C. L. Chien, and N. Markovic for sharing their unpublished data. This work was supported in part by NSF Grant DMR-0520491.

<sup>1</sup> J. E. Davies, O. Hellwig, E. E. Fullerton, G. Denbeaux, J. B. Kortright, and K. Liu, Phys. Rev. B **70**, 224434

(2004).

- <sup>2</sup> L. B. Steren, J. Milano, V. Garcia, M. Marangolo, M. Ed-drief, and V. H. Etgens, Phys. Rev. B **74**, 144402 (2006).
- <sup>3</sup> O. Donzelli, D. Palmeri, L. Musa, F. Casoli, F. Albertini, L. Pareti, and G. Turilli, J. Appl. Phys. **93**, 9908 (2003).
- <sup>4</sup> Y. L. Iunin, Y. P. Kabanov, V. I. Nikitenko, X. M. Cheng, D. Clarke, O. A. Tretiakov, O. Tchernyshyov, A. J. Sharpiro, R. D. Shull, and C. L. Chien, Phys. Rev. Lett. **98**, 117204 (2007).
- <sup>5</sup> A. Berger and R. P. Erickson, J. Magn. Magn. Mater. **165**, 70 (1997).
- <sup>6</sup> A. B. Kashuba and V. L. Pokrovsky, Phys. Rev. B **48**, 10335 (1993).
- <sup>7</sup> Ar. Abanov, V. Kalatsky, V. L. Pokrovsky, and W. M. Saslow, Phys. Rev. B **51**, 1023 (1995).
- <sup>8</sup> T. Garel and S. Doniach, Phys. Rev. B **26**, 325 (1982).
- <sup>9</sup> Y. Yafet and E. M. Gyorgy, Phys. Rev. B **38**, 9145 (1988).
- <sup>10</sup> D. Clarke, O. A. Tretiakov, and O. Tchernyshyov, Phys. Rev. B (To be published) cond-mat/0612346
- <sup>11</sup> S. H. Lee, F. Q. Zhu, C. L. Chien, and N. Markovic, unpublished.
- <sup>12</sup> A. Marty, Y. Samson, B. Gilles, M. Belakhovsky, E. Dudzik, H. Durr, S. S. Dhesi, G. van der Laan, and J. B. Goedkoop, J. Appl. Phys. **87**, 5472 (2000).
- <sup>13</sup> A. L. Sukstanskii, and K. I. Primak, J. Magn. Magn. Mater. **169**, 31 (1997).
- <sup>14</sup> R. C. O'Handley, *Modern Magnetic Materials: Principles and Applications* (Wiley & Sons, New York, 2000) p. 655.
- <sup>15</sup> O. Schulte, F. Klose, and W. Felsch, Phys. Rev. B **52**, 6480 (1995).